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Formation and Thickness Evolution of Periodic Twin Domains in Manganite Films Grown on $\text{SrTiO}_3(001)$ Substrates

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We reveal a new kind of misfit strain relaxation process in the growth of thin manganite films on $\text{SrTiO}_3(001)$ substrates that exploits twinning to adjust lattice mismatch. We show that this relaxation mechanism emerges in thin films as one-dimensional twinning waves, which freeze out into a twin domain pattern as the manganite film continues to grow. A quantitative microscopic model that uses a matrix formalism is able to reproduce all x-ray features and provides a detailed insight into this novel relaxation mechanism. We further demonstrate how this twin angle pattern affects the transport properties in these functional films.

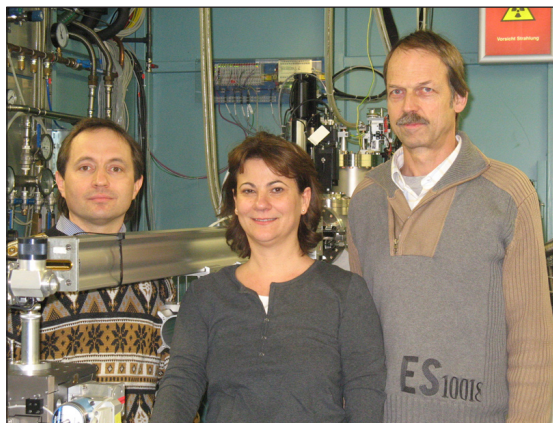
In bulk manganese oxides, the transport properties depend on band filling (doping degree), temperature, magnetic field, and eventually present lattice distortions. Since future applications will extensively exploit thin-film geometries, the associated transport properties will strongly be determined by the strain misfit as caused by the microscopic clamping of the film to the substrate. For example, Sr-doped lanthanum manganite films $\text{La}_{1-\delta}\text{Sr}_\delta\text{MnO}_3$ (denoted as LSMO) ($\delta = 0.10$, $t \leq 50$ nm) grown on an $\text{SrTiO}_3(001)$ substrate (denoted as STO) show metallic behavior at low temperatures instead of the expected insulating bulk behavior. Thus, by a microscopic understanding of this misfit strain relaxation mechanism, one may get a tool at hand for tuning the electronic properties of manganite films.

Here we report an x-ray diffraction study of the strain relaxation behavior of LSMO films with compositions $\delta = 0.10$, 0.125 and with film

thicknesses varying from 12 to 110 nm, which have been grown on STO by using pulsed laser deposition. They experience compressive as well as tensile strain ranging from -0.45% to +0.95% lattice mismatch, respectively. We will show that these films exhibit an intriguing strain accommodation scenario: they first develop periodic one-dimensional (1D) twinning modulation waves, which progressively develop into a twin domain (TD) pattern as the film thickness increases.

The average structure of all investigated LSMO films is pseudomorphic with respect to the STO substrate. **Figure 1** shows two data sets displaying the intensity distributions in the $[0\eta 0]$ direction for the 26 nm and 88 nm LSMO films representing the thin- and thick-film situation. Our key observations are as follows: (a) The thin-film scenario (**Figure 1a**) is dominated by superstructure satellite peaks that emerge with a constant in-plane momentum transfer $\Delta\eta$ implying a periodic height modulation $z(y)$

with a periodicity $2d$. The observed increase of the FWHM of the satellite peaks with $|\eta|$ discloses that this in-plane modulation is short ranged. (b) The thick-film scenario (**Figure 1b**) is dominated by twin peaks whose lateral η position increases linearly with L value. These twin peaks originate from individual TDs with (001) lattice planes tilted by an angle Φ_z with respect to the surface. These two dominating structural motifs are illustrated in **Figure 2a**, the modulation structure from periodic twin-



Authors (from left), Nikolai Kasper, Assunta Vigliante, and Peter Wochner

ning (left) and laterally coherent tilt domains (right). Notice that both motifs have the same origin: tilted lattice planes. Satellite peaks and twin peaks are simultaneously observed, especially well in the thick film (see red lines in **Figure 1b**). This is a strong indication that a 1D-modulated structure of coherently connected TDs is formed. For a quantitative microscopic modeling of these strain relaxation phenomena we have developed a statistical matrix description, which is closely connected with a model conceived for the description of surface faceting. As can be seen from the fit curves in **Figure 1**, our

model describes all salient features in a self-consistent and quantitative way.

Figure 2b summarizes the measured temperature dependence of the (004) x-ray peak maxima associated with the 26 nm film (see **Figure 1**) together with the recorded electrical resistance. We observe, that the satellite peaks disappear in accordance with resistivity below $T = 220$ K. This is the signature of a triclinic (related to $R\bar{3}c$) to monoclinic (related to $Pbnm$) phase transition in which z disappears abruptly. Because of the strain gradient along z within the

film, the structural phase transition is broadened between 100 and 220 K. This triclinic-monoclinic phase transition affects in turn strongly the octahedral MnO_6 tilt system and, therefore, establishes a strong correlation between octahedral tilts and twin modulation waves.

In summary, we have unraveled a novel strain relaxation phenomenon, which exploits the delicate balance between MnO_6 octahedral tilts and the formation of twin modulation waves to adjust the manganite lattice parameter to the substrate.

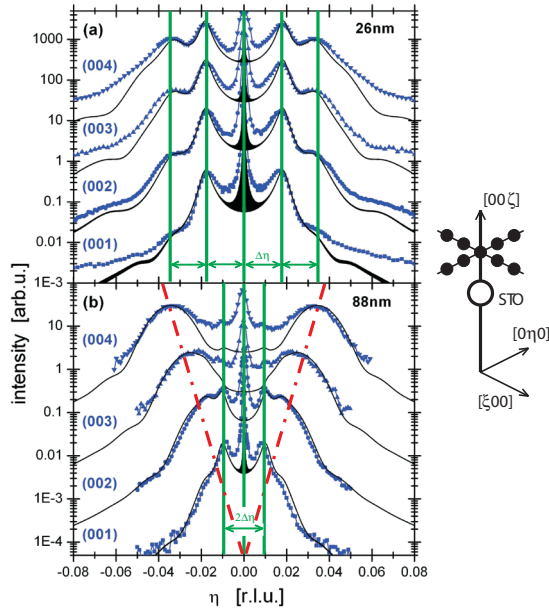


Figure 1. (a) Transverse scans of a 26 nm film at (001) to (004) at RT showing dominantly central peak and 1st and 2nd order satellite peaks (green solid lines). (b) transverse scans of a 88 nm film at (001) to (004) at RT showing dominantly twin peaks (red dashed lines) as a guide to the eye) due to the tilted lattice planes. The numerically calculated intensity distribution of each reflection (solid curve) is superimposed using the microscopic matrix model. The sketch on the right shows the (00L) associated peak positions of the film (full circles) and the substrate (open circle), respectively.

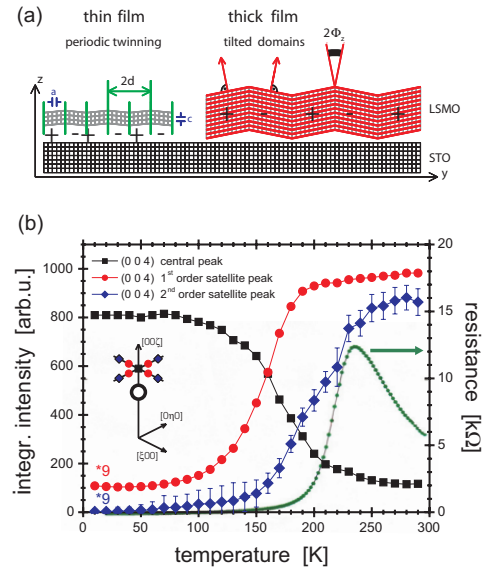


Figure 2. (a) Real-space sketch to show which motifs of the film structure, grown on average pseudomorphically on top of a rigid, cubic substrate (STO), are dominant in the case of a thin (left) and a thick manganite film (right). The difference in variance of the TD length for the two cases is not illustrated in the picture. (b) Temperature dependence of the structure (integrated intensity of the central peak and the 1st and 2nd order satellite peaks of the (004) Bragg reflection, whose η positions are indicated by vertical green lines in **Figure 1a**) and transport of the 26 nm LSMO film. The inset shows the (00L) associated film (solid symbols) and substrate (open circle) peak positions.